



Analysis of fuel element matrix graphite corrosion in HTR-PM for normal operating conditions

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ARTICLE INFO

Article history:

Received 2 June 2009

Received in revised form

16 November 2009

Accepted 10 December 2009

ABSTRACT

The corrosion of spherical fuel element by oxidizing gases will degrade their thermal and mechanical properties in pebble-bed reactors. Oxidizing impurities in primary helium coolant may have significant influence on the gasification of graphite during the long service time even though their concentrations remain relatively low. This paper deals with the corrosion of matrix graphite by steam and oxygen in Chinese high temperature gas-cooled reactor pebble-bed module (HTR-PM). The influence of steam contents was first analyzed, and then the effect of oxygen contents was also taken into account. The results show that the corrosion by steam was weak and it would not threaten the normal service of spherical fuel elements for expected steam contamination levels. On the contrary, the corrosion would be more severe while the oxygen content was as high as currently expected. Finally, the upper limits of steam and oxygen in primary helium coolant were recommended to be 1.0 and 0.05 cm³ m⁻³.

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1. Introduction

China began research work on pebble-bed high temperature gas-cooled reactors at the end of 1970s and the HTR-10 project has been successfully designed and built at the Institute of Nuclear and New Energy Technology (INET) of Tsinghua University. On the basis of the success of HTR-10, the high temperature gas-cooled reactor pebble-bed modular (HTR-PM) is proposed, which aims to demonstrate the claimed inherent safety features and its economic competitiveness (Zhang and Sun, 2007). The HTR-PM project will surely be benefit from the rich design and construction experiences of HTR-10.

The HTR-PM adopted spherical fuel element, which is largely made up of matrix material which is made from natural graphite, electro graphite and phenol resin. The matrix graphite has good mechanical properties and good corrosion resistance. However, the matrix graphite may be corroded severely when oxidizing gases enters into the core with large quantities for air or water ingress accidents. Many authors have studied the corrosion of matrix graphite for accidents conditions and showed that the spherical fuel element would keep its integrity since the core would be cooled effectively. But in fact the matrix graphite will also be gasified slowly by the impurities in helium coolant for normal operating conditions, which can be accelerated by the high temperature in

reactor core (Gastaldi et al., 2006). Eto et al. (1986) and Richards (1990) investigated the corrosion of fuel element graphite in block type reactors for certain temperatures and obtained the burn-off at low steam contents. Hinssen et al. (2008) reviewed the background concerning graphite oxidation and Kim et al. (2008) studied the influence of steam on graphite oxidation by oxygen, which showed that the steam did not affect the reaction rate for temperature lower than 1170 K.

Though the corrosion of spherical fuel element is very slow for normal operating conditions, this slow corrosion will actually also be accumulated for long time and may cause significant degradation of the mechanical properties of fuel element, which may accelerate the production of graphite dust in consequence. Here, this paper mainly describes the estimation of matrix graphite corrosion of spherical fuel element and recommends the upper limits of oxidizing gases in primary helium coolant of HTR-PM for normal operating conditions.

2. Brief description of HTR-PM

The HTR-PM project has two reactors, which are arranged side by side. The reactor core is pebble-bed like and is surrounded by the reflector piled with quantities of graphite bricks. There are about 420 000 spherical fuel elements in one module of HTR-PM. The following parameters of HTR-PM are for one module if there is no particular identification. The reactor core of HTR-PM has a diameter of 3 m and an average height of 11 m. The spherical fuel elements are packed disorderly in the core with a mean packing density of

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0.61. The spherical fuel element in HTR-PM has the same structure with the fuel element in HTR-10, which is 60 mm in diameter and is coated with a 5 mm graphite layer that is fuel free. The tripe coated (TRISO) type fuel particles are homogeneously dispersed in the graphite matrix of the fuel zone. The coated particles in each fuel element will increase from 8300 to about 12 000 compared to HTR-10 (Zhao et al., 2006), so the heavy metal loading will reach about 7 g.

The HTR-PM core adopts a continuous and multi-pass loading mode, and every sphere will be circulated 15 times on average in the core. There will be 6000 spherical fuel elements recycled every day, in which 379 fresh fuel elements will be added and the same number of waste fuel elements will be taken off. So the mean residence time of spherical fuel element is around 1057 days in HTR-PM core. The primary pressure of the HTR-PM is 7.0 MPa and the thermal power is 250 MW for every module. The helium temperature at the inlet and the outlet of the core are 250 °C and 750 °C, respectively. The helium coolant flows into the reactor core via an annular gap between the hot duct and the cross vessel. Then, it goes downwards and changes its direction at the lower part of the reactor pressure vessel. Most of the helium flows to the cold helium plenum at the upper part of the top reflector through the cold helium channels in the graphite blocks of the side reflector. The mainstream helium passes through the reactor core to cool effectively the fuel elements. Then the high temperature helium is mixed sufficiently in the hot helium plenum and is transported to the power conversion system through the hot duct.

3. Simplifications to the HTR-PM core

The flow of helium is really complicated in HTR-PM core because of its complex structure, especially the piled spherical fuel elements and graphite balls which induces the distortion of the flow field. This has brought more difficulties to the simulations of helium flow in the pebble-bed core and also to the estimation of graphite corrosion. The pebble-bed core was mainly simplified to porous media to study the helium flow and the corrosion of graphite for accident conditions (Gao and Wang, 1993; Becker and Laurien, 2003; Walter et al., 2006; Studer et al., 2007). There are also researchers who investigate the helium flow between spherical fuel elements with direct numerical simulations, but this method is too computationally intensive. Though some researchers have investigated the flow and heat transfer between several dozens of spherical fuel elements (Siu and Lee, 2004; Lee et al., 2007; Hassan, 2008), it is still too difficult to simulate the whole core with direct method yet. Therefore, an equivalent cylindrical model was employed to simplify the real structure during estimating the matrix graphite corrosion in pebble-bed reactors. The main principle of simplification was the constant of the spherical fuel elements volume. It was also required that the outer surface area of graphite material in the simplified model was equal to the surface of all the spheres in the pebble-bed core. Thus, the internal diameter of the equivalent cylindrical model was obtained as 25.6 mm.

The mean residence time of spherical fuel element is about 1057 days in HTR-PM core and every sphere will cross the core 15 times on average, so it takes about 70 days for a fuel element moving from the top to the bottom of the core. The pebble's flow is quite complicate in HTR-PM core because of its randomness. But the path lines of fuel elements also show regular appearance in macroscopic view (Hu et al., 2009). The closer the pebble is to the bottom, the faster the velocity is. As a preliminary study, the spherical fuel element was simplified to move uniformly in the core in this simulation. The influence of pebble's location on its speed was neglected and the moving path was equivalent to the height of the reactor core. Therefore, the speed of fuel element was expressed as $v_{SF} = H/T_c$,

where v_{SF} is the speed; H is the height of reactor core and T_c is the mean cycle time of fuel element.

The corrosion of matrix graphite would change its burn-off, which represents the ratio of consumed graphite to the initial mass, and accelerate the oxidation rate, so the burn-off factor should be taken into account when simulating the oxidation of spherical fuel elements. The temperature and burn-off of fuel element matrix graphite would vary with its location in HTR-PM core. The temperature of fuel element could be represented by the time integral of pebble's speed. So the burn-off of fuel free zone was expressed as following for every cycle,

$$B_N = \int_0^{T_c} \frac{MR}{M_{FF}} dv dt \quad (1)$$

where B_N is the burn-off of fuel free zone of spherical fuel element; R is the corrosion rate of matrix graphite; M is the molar mass of graphite and M_{FF} is the mass of fuel free zone.

The spherical fuel element of HTR-PM would be circulated for 15 times on average and the burn-off of graphite at the end of one cycle would be the initial condition for the following cycle. Therefore, the burn-off of fuel free zone of the spherical fuel element at the end of life time could be calculated by

$$B_{SFF} = \sum_{N=1}^{15} \int_{(N-1)T_c}^{NT_c} \frac{MR}{M_{FF}} dv dt \quad (2)$$

where B_{SFF} is the total burn-off of matrix graphite at the end of life time and N is the number of cycles.

The corrosion rate of matrix graphite was much slower in HTR-PM for normal operating conditions because of the very low contents of oxidizing gases, which might come from the contaminants in new helium supply, degassing of graphite materials and internal metallic structure and the working operation (Yao et al., 2002; Legros et al., 2006). The upper limits of gaseous impurities in primary helium coolant are shown in Table 1, which referred to the HTR-10 values. The impurities limits shown in Table 1 seem higher than the values of High Temperature Engineering Test Reactor (HTR) presented by Sakaba and Hirayama (2005), especially for the steam and oxygen which were 0.2 and 0.04 cm³ m⁻³ in HTR. But in fact, the helium purification system would eliminate effectively the impurities in helium for normal operating conditions and ensure that the impurities contents could satisfy the requirements of the core. For instance, the steam content of HTR-10 was reported as below 0.33 cm³ m⁻³ for normal operating conditions (Luo and Yu, 2007). Thus, the low corrosion rate of matrix graphite would only produce negligible heat emission, which would not influence the temperature distribution in the core for normal operating conditions. So the steady temperature distribution was adopted in simulating the corrosion of fuel element.

The large part of spherical fuel element in HTR-PM was matrix graphite other than the standard nuclear graphite in block type reactor. Moreover, the matrix graphite was suspected sometimes to be less corrosion resistance. Moormann et al. (2004) compared the oxidation behavior of matrix graphite with standard nuclear

Table 1
Upper limits of impurities contents in primary helium coolant of HTR-PM.

Impurities	Contents (cm ³ m ⁻³)
H ₂ O	2
H ₂	30
CO	30
O ₂	0.2
N ₂	2
CO ₂	6
CH ₄	5

graphite in oxygen and concluded that their burn-off behaviors were quite similar. Kuhn et al. (2004) also indicated that the overall reaction rate of matrix graphite was not higher compared with standard nuclear graphite. Thus, the kinetic parameters of standard nuclear graphite were applied in the simulations.

4. Simulations and discussions

4.1. The steam corrosion

The upper limit of steam is much higher than oxygen in primary helium and the steam corrosion to matrix graphite was also considered to be more important in HTGR for normal operating conditions. So, the sole steam corrosion, which produced carbon monoxide and hydrogen, was first simulated in HTR-PM. The conservation equation of steam in matrix graphite is shown as below,

$$\frac{\partial C_{H_2O}}{\partial t} + \nabla(-D_{e-H_2O} \nabla C_{H_2O}) = -R_{H_2O} \quad (3)$$

where C_{H_2O} denotes steam concentration, D_{e-H_2O} the effective diffusion coefficient and R_{H_2O} the reaction rate. The Langmuir–Hinshelwood equation was used to describe the reaction, which is shown as,

$$R_{H_2O} = \frac{\rho F_b K_1 P_{H_2O}}{M(1 + K_2 P_{H_2}^\beta + K_3 P_{H_2O})} \quad (4)$$

and the kinetic parameters, K_1 , K_2 and K_3 were given in (Velasquez et al., 1978). ρ and M are the graphite density and molar mass of carbon. P_{H_2O} and P_{H_2} are the partial pressure of steam and hydrogen. The effect of burn-off on reaction rate, F_b , was expressed by Eq. (5) (Bhatia and Perlmutter, 1980):

$$F_b = (1 - X) [1 - \xi_0 \ln(1 - X)]^{0.5} \quad (5)$$

In Eq. (5), F_b is the burn-off factor, X the burn-off and ξ_0 is the structure parameter, which was taken as 45 in calculations. The burn-off of graphite was calculated by

$$\frac{\partial X}{\partial t} = \frac{MR_{H_2O}}{\rho_{g0}} \quad (6)$$

where ρ_{g0} was the initial density of graphite.

Fig. 1 shows the influence of steam pressure on the corrosion of the fuel free zone of fuel element, where the hydrogen content was assumed as $30 \text{ cm}^3 \text{ m}^{-3}$. It is shown that the corroded graphite increased quickly with increasing steam partial pressure, and the burn-off reached about 2.39% at steam content of $2.0 \text{ cm}^3 \text{ m}^{-3}$. The

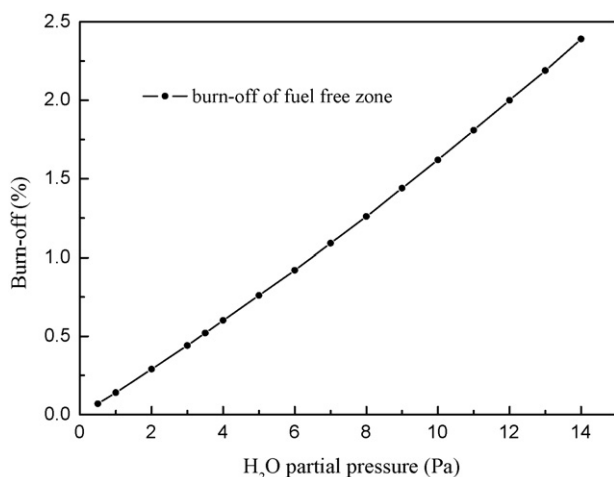


Fig. 1. The influence of H_2O partial pressure on graphite corrosion in HTR-PM.

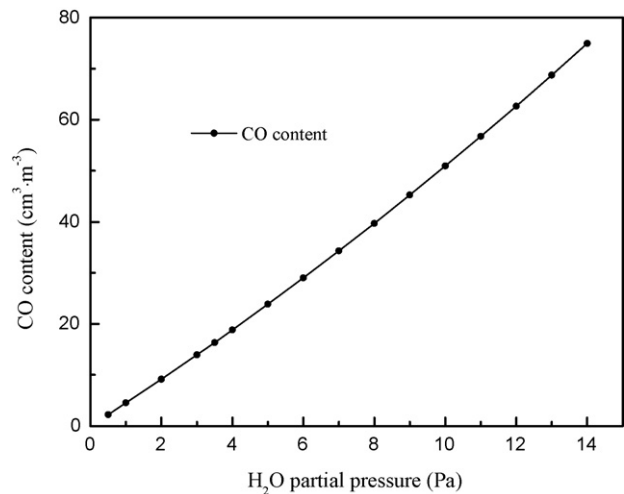


Fig. 2. The CO content caused by steam corrosion in HTR-PM.

designed fuel elements was around 420 000 in the HTR-PM core, so the consumed matrix graphite of all the fuel elements would be as high as 280 kg every year. The total mass would reduce to 130 and 60 kg per year for steam contents of 1.0 and $0.5 \text{ cm}^3 \text{ m}^{-3}$, respectively. It seemed that the corroded graphite of fuel elements in HTR-PM core was large, which was as high as several hundred kilogram for high steam content. Therefore, the steam content in helium coolant should be controlled strictly.

The total consumed matrix graphite in HTR-PM was much higher than in HTR-10 for the same volume contents of steam. The primary helium pressure in HTR-PM was higher than HTR-10, which resulted in high steam partial pressure for the same volume content. Eq. (4) showed that the reaction rate depended on the steam partial pressure, so the corrosion rate of matrix graphite in HTR-PM was faster than in HTR-10. In addition, the fuel elements in HTR-PM were much more than in HTR-10, which also led to more graphite corroded. So, the higher corroded graphite did not mean more severe oxidizing condition. In fact, the burn-offs of graphite were similar for the same molar concentration of steam.

The helium purification system of HTR-PM was designed for a helium flow rate of 150 kg h^{-1} , corresponding with a 5% of the helium inventory in primary circuit. Fig. 2 shows the contents of carbon monoxide caused by steam corrosion when the production rates of hydrogen and carbon monoxide reached quasi-equilibrium state with the purification capacity of helium purification system. The contents of carbon monoxide increased quickly with increasing steam partial pressure. The contents of carbon monoxide caused by steam corrosion were 16, 34 and $75 \text{ cm}^3 \text{ m}^{-3}$ for steam content of 0.5, 1.0 and $2.0 \text{ cm}^3 \text{ m}^{-3}$. However, the upper limit of carbon monoxide in HTR-PM core was required below $30 \text{ cm}^3 \text{ m}^{-3}$. Thus, the steam content in primary helium coolant should below $1.0 \text{ cm}^3 \text{ m}^{-3}$ to restrain the contents of hydrogen and carbon monoxide, where the corrosion rate of all the fuel elements was about 1.2 mol h^{-1} for this circumstance.

The chemical reaction of steam and fuel element matrix graphite would degrade the mechanical properties of fuel element. The slight increase of wearing extent of every pebble would result in the production of large amount of dusty because of the huge fuel elements in HTR-PM core. Fig. 3 displays the burn-off of matrix graphite at steam content of $1.0 \text{ cm}^3 \text{ m}^{-3}$. The burn-off of matrix graphite was quite low and showed a uniform distribution with depth into graphite. The corrosion rate of matrix graphite was mainly controlled by the very low chemical reaction rate for normal operating condition, so the steam could diffuse into the matrix graphite and react with graphite uniformly. The maximum burn-

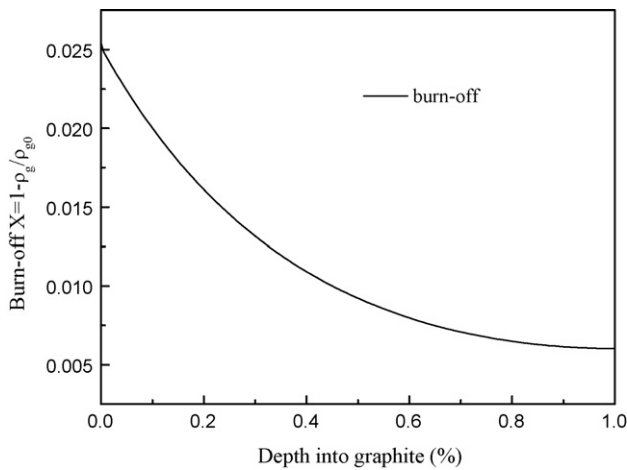


Fig. 3. The burn-off of fuel element at H_2O content of $1.0 \text{ cm}^3 \text{ m}^{-3}$.

off of fuel element was about 2.5% at steam content of $1.0 \text{ cm}^3 \text{ m}^{-3}$, which had very slight influence on fuel element. However, the corrosion of fuel element shown in Fig. 4 at steam content of $2.0 \text{ cm}^3 \text{ m}^{-3}$ was much more severe. The maximum burn-off was about 6.0%, which might have important consequence on the fuel element. Therefore, the steam content should be restrained at low level to prevent the decrease of wear property of fuel element. It was reasonable to restrict the steam content below $1.0 \text{ cm}^3 \text{ m}^{-3}$ in primary helium loop from Figs. 3 and 4.

The weight loss of fuel element could be calculated by integrating the consumed graphite of every cycle, which was expressed as following,

$$B_{SF} = \sum_{N=1}^{15} \int_{(N-1)T_c}^{NT_c} \frac{\rho_{H_2O} M_{R_{H_2O}}}{M_{SF}} dv dt \quad (7)$$

where B_{SF} and M_{SF} are the weight loss and the mass of fuel element. The variation of weigh loss of fuel element at steam content of $1.0 \text{ cm}^3 \text{ m}^{-3}$ is given in Fig. 5. It shows that the mean weight loss of fuel element was about 0.45% at the end of service time. In other words, approximately 0.05 mm of graphite was exhausted. The weight loss of fuel element would increase to 0.97% at steam content of $2.0 \text{ cm}^3 \text{ m}^{-3}$. On the contrary, the weight loss reduced to only 0.21% at steam content of $0.5 \text{ cm}^3 \text{ m}^{-3}$.

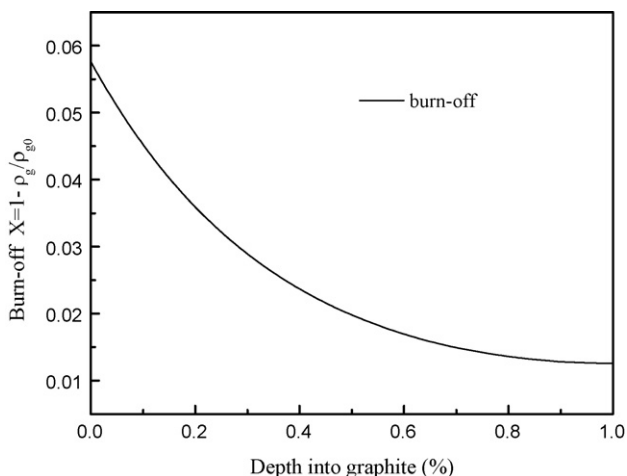


Fig. 4. The burn-off of fuel element at H_2O content of $2.0 \text{ cm}^3 \text{ m}^{-3}$.

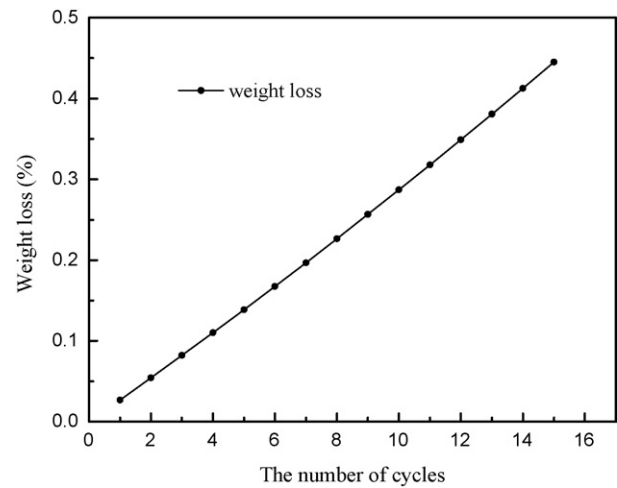


Fig. 5. The weight loss of fuel element with the number of cycles at H_2O content of $1.0 \text{ cm}^3 \text{ m}^{-3}$.

4.2. The influence of oxygen

Though the content of oxygen is very low in HTR-PM, the reaction rate of graphite and oxygen is much higher than steam, which may result in much more serious corrosion when the oxygen content increases. Here, the influence of oxygen was studied for higher oxygen contents when the steam content was $1.0 \text{ cm}^3 \text{ m}^{-3}$. It should be noted that the reduction of oxygen content was ignored during pass through the cold helium channels in the side reflector because of its low temperature, and the corrosion to the bottom reflector was not studied here. The conservation of oxygen in matrix graphite is given below:

$$\frac{\partial C_{O_2}}{\partial t} + \nabla \cdot (-D_{e-O_2} \nabla C_{O_2}) = -R_{O_2} \quad (8)$$

where C_{O_2} is the concentration of oxygen, D_{e-O_2} the effective diffusion coefficient of oxygen in matrix graphite and R_{O_2} the reaction rate of oxygen and graphite. The reaction rate of oxygen and graphite was expressed by power-law equation, $R_{O_2} = 0.452 \exp(-19071/T) P_{O_2} F_b$ (Luo and Yu, 2007), where the P_{O_2} is the partial pressure of oxygen. The corrosion rate of graphite relied on the coupling effect of steam and oxygen, so the burn-off of matrix graphite could be obtained by Eq. (9):

$$\frac{\partial X}{\partial t} = \frac{M(R_{H_2O} + R_{O_2})}{\rho_{g0}} \quad (9)$$

Fig. 6 shows the burn-off of fuel element for oxygen contents in the range of $0-0.2 \text{ cm}^3 \text{ m}^{-3}$. The burn-off of matrix graphite of the fuel free zone increased with increasing oxygen partial pressure, and this increase slowed down slightly at high oxygen contents. Moreover, the burn-off was higher than considering only the steam because of the fast chemical reaction rate of oxygen and graphite. The burn-off of the fuel free zone was about 3.88% at oxygen content of $0.1 \text{ cm}^3 \text{ m}^{-3}$ (or 0.7 Pa) and the consumed matrix graphite could reach about 420 kg every year. The burn-off of matrix graphite would reduce from 3.88% to 2.52% while the oxygen content reduced by half and the consumed graphite would also reduce to 300 kg every year. Therefore, it could be concluded that the influence of oxygen in helium coolant was very important.

Fig. 7 gives the profile of matrix graphite burn-off at oxygen contents of 0.1 and $0.05 \text{ cm}^3 \text{ m}^{-3}$. The burn-off of matrix graphite caused by oxygen and steam was much higher than that caused by steam solely. Furthermore, the corrosion was mainly occurred near the surface and the burn-off decreased very sharply with the depth into graphite. It was indicated previously that the reaction

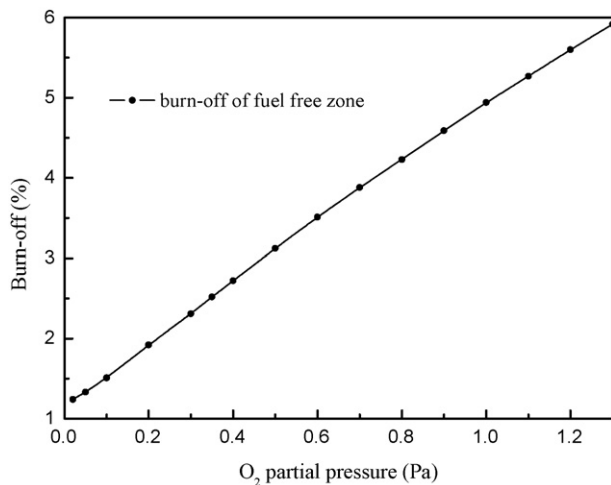


Fig. 6. The influence of O_2 content on graphite corrosion in HTR-PM.

rate of oxygen and fuel element matrix graphite was much rapid and the corrosion was mainly controlled by the diffusion rate of oxygen. Most of the oxygen was consumed near the surface of fuel element and could not diffuse into matrix graphite. Fig. 7 also shows that the corrosion at oxygen content of $0.1 \text{ cm}^3 \text{ m}^{-3}$ was more severe than of $0.05 \text{ cm}^3 \text{ m}^{-3}$. The corrosion was restricted to about 10% of the depth of graphite at oxygen content of $0.1 \text{ cm}^3 \text{ m}^{-3}$ and 0.1 mm of graphite was exhausted near the surface. But the main corrosion region was restricted to about 5% of the depth and only 0.05 mm of matrix graphite was exhausted at oxygen content of $0.05 \text{ cm}^3 \text{ m}^{-3}$. The weight loss of fuel element by oxygen and steam could also be obtained by Eq. (7), where the R_{H_2O} was replaced by $(R_{H_2O} + R_{O_2})$, and the trends of weight loss with the number of cycles were given in Fig. 8. The weight loss increased very quickly with the cycles and this increase was also accelerated slowly. In addition, the increase rate at oxygen content of $0.1 \text{ cm}^3 \text{ m}^{-3}$ was higher than of $0.05 \text{ cm}^3 \text{ m}^{-3}$. The weight loss of fuel element at the end of service time reached about 1.58% at oxygen content of $0.1 \text{ cm}^3 \text{ m}^{-3}$ and steam content of $1.0 \text{ cm}^3 \text{ m}^{-3}$ for normal operating condition, which also meant that 0.16 mm of graphite was exhausted. The weight loss would reduce to 1.03% at oxygen content of $0.05 \text{ cm}^3 \text{ m}^{-3}$ and only 0.11 mm of graphite would be exhausted.

It can be seen that the corrosion of fuel element at oxygen of $0.1 \text{ cm}^3 \text{ m}^{-3}$ was much more severe than of $0.05 \text{ cm}^3 \text{ m}^{-3}$. More-

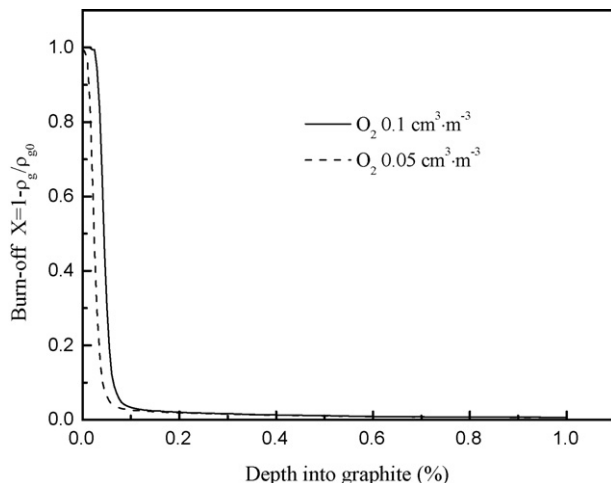


Fig. 7. The burn-off of fuel element for different O_2 contents.

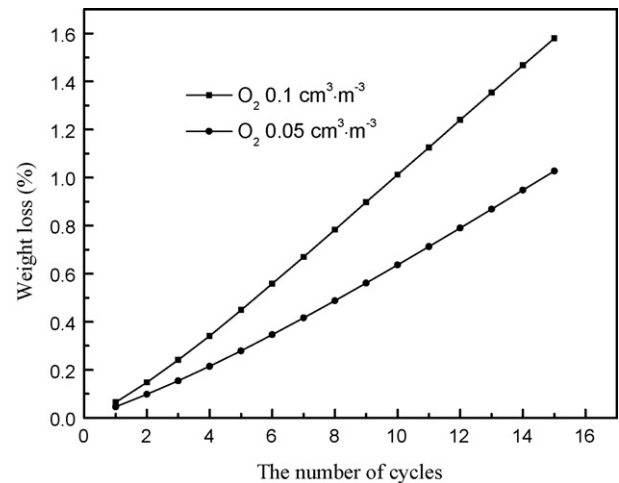


Fig. 8. The weight loss of fuel element with the number of cycles for different O_2 contents.

over, the surface of fuel element was corroded critically because of the oxygen, which might degrade the properties of matrix graphite significantly. So the oxygen content was recommended as below $0.05 \text{ cm}^3 \text{ m}^{-3}$ to prevent the corrosion of fuel element and the production of dusty for normal operating conditions.

5. Conclusions

The corrosion behaviors of fuel element matrix graphite were simulated in the paper for normal operating condition. The amount of consumed graphite increased very quickly with increasing steam content. The consumed graphite could reach about 280 kg per year at steam content of $2.0 \text{ cm}^3 \text{ m}^{-3}$ in helium coolant, and the maximum burn-off of matrix graphite was about 6%. Contrarily, the total amount of consumed graphite reduced to 130 kg per year at steam content of $1.0 \text{ cm}^3 \text{ m}^{-3}$. Though the amount of consumed graphite in HTR-PM was huge, it did not mean much severe corrosion occurred in HTR-PM because of the large quantity of matrix graphite.

The burn-off of matrix graphite was so low that it would not affect the geometry of spherical fuel element. Additionally, the mean weight loss of fuel element was about 0.45% at steam content of $1.0 \text{ cm}^3 \text{ m}^{-3}$, or in other word, approximate 0.05 mm of matrix graphite was exhausted on the surface. In brief, the steam corrosion would not affect critically the service of fuel element in HTR-PM.

The corrosion to fuel element by oxygen and steam was much more severe than only taking into account the steam. The consumed graphite of all the fuel elements was even 420 kg per year at steam and oxygen contents of 1.0 and $0.1 \text{ cm}^3 \text{ m}^{-3}$. Moreover, the chemical reaction rate of oxygen and graphite was quite rapid so that the corrosion to matrix graphite was restricted to about 10% of the depth to the surface. The weight loss of fuel element was about 1.58% for such an atmosphere, but the weight loss would reduce to 1.03% when the oxygen content reduced by half. Therefore, the oxygen content should be controlled strictly to avoid the corrosion of fuel element matrix, especially the presence of oxygen during the fuel loading operations for normal operating conditions. It was finally recommended that the contents of steam and oxygen should be controlled below 1.0 and $0.05 \text{ cm}^3 \text{ m}^{-3}$.

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